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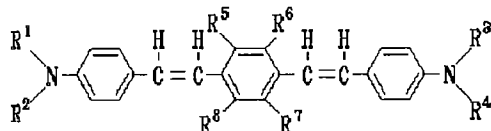
(54) 【発明の名称】 有機電界発光素子

(57) 【要約】 (修正有)

【課題】 安定かつ発光輝度の大きい赤色発光の有機電界発光素子を提供すること。

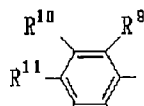
【解決手段】 発光領域を有する有機層が陽極と陰極との間に設けられている有機電界発光素子において、有機層に下記一般式(1)で表されるジスチリル化合物が含まれていることを特徴とする、有機電界発光素子。

一般式(1)：



〔但し、前記一般式(1)において、R¹、R²、R³及びR⁴は互いに同一の又は異なる基であって、下記一般式(2)で表わされるアリール基であり

一般式(2)：

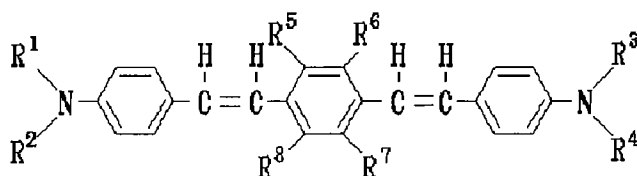


(但し、前記一般式(2)において、R⁹、R¹⁰、R¹¹、R¹²、R¹³は互いに同一の又は異なり、その少なくとも1つが飽和又は不飽和アルコキシ基、又はアルキル基である。)、R⁵、R⁶、R⁷、R⁸は互いに同一の又は異なる基であって、それらの少なくとも1つがシアノ基、ニトロ基又はハロゲン原子である。〕

【特許請求の範囲】

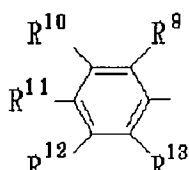
【請求項 1】 発光領域を有する有機層が陽極と陰極との間に設けられている有機電界発光素子において、前記有機層に下記一般式（1）で表されるジスチリル化合物

一般式（1）：



〔但し、前記一般式（1）において、R¹、R²、R³及びR⁴は互いに同一の又は異なる基であって、下記一般式（2）で表わされるアリール基であり

一般式（2）：



（但し、前記一般式（2）において、R⁹、R¹⁰、R¹¹、R¹²及びR¹³は互いに同一の又は異なる基であって、それらの少なくとも1つが飽和又は不飽和アルコキシル基、又はアルキル基である。）、R⁵、R⁶、R⁷及びR⁸は互いに同一の又は異なる基であって、それらの少なくとも1つがシアノ基、ニトロ基又はハロゲン原子である。）

【請求項 2】 前記有機層が、正孔輸送層と電子輸送層とが積層された有機積層構造を有しており、前記正孔輸

送層の形成材料として前記ジスチリル化合物が用いられている、請求項 1 に記載した有機電界発光素子。

【化 1】

送層の形成材料として前記ジスチリル化合物が用いられている、請求項 1 に記載した有機電界発光素子。

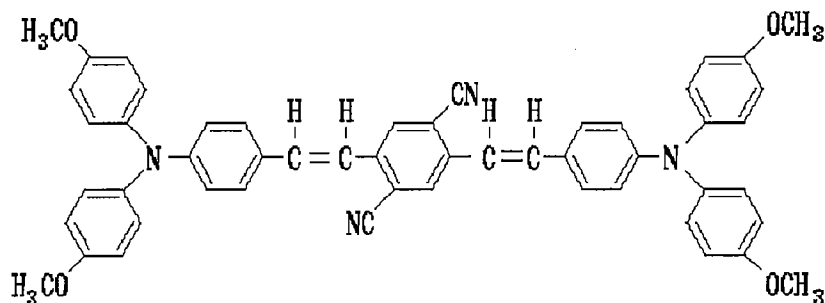
【請求項 3】 前記有機層が、正孔輸送層と電子輸送層とが順次積層された有機積層構造を有しており、前記電子輸送層の形成材料として前記ジスチリル化合物が用いられている、請求項 1 に記載した有機電界発光素子。

【請求項 4】 前記有機層が、正孔輸送層と発光層と電子輸送層とが積層された有機積層構造を有しており、前記発光層の形成材料として前記ジスチリル化合物が用いられている、請求項 1 に記載した有機電界発光素子。

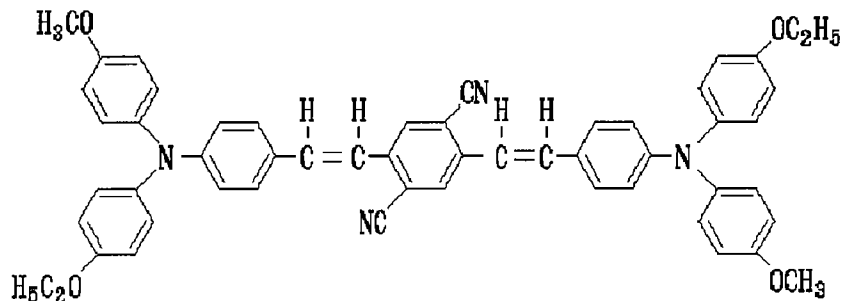
【請求項 5】 発光領域を有する有機層が陽極と陰極との間に設けられている有機電界発光素子において、前記有機層に下記構造式（3）- 1、（3）- 2、（3）- 3、（3）- 4、（3）- 5、（3）- 6 又は（3）- 7 で表されるジスチリル化合物の少なくとも 1 種が有機発光材料として含まれていることを特徴とする、有機電界発光素子。

【化 2】

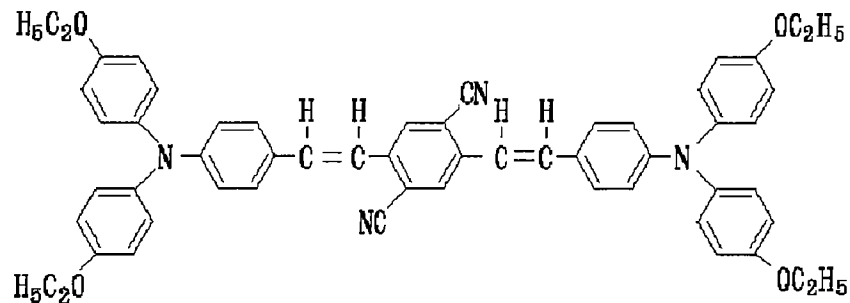
構造式（3）- 1：



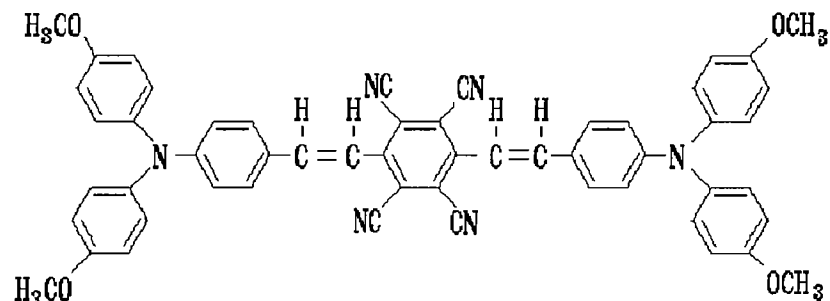
構造式（3）- 2：



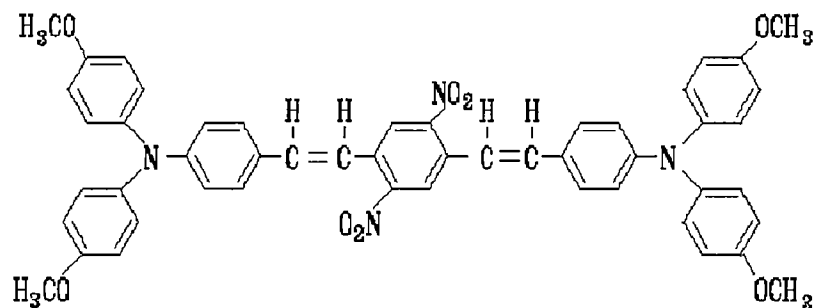
構造式 (3) - 3 :



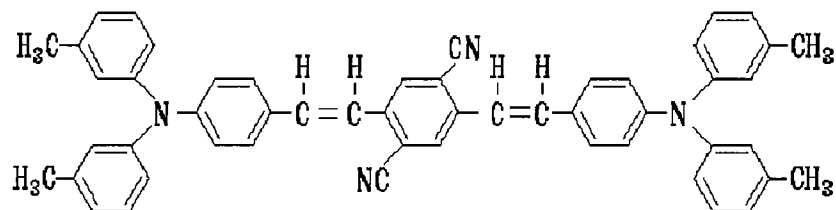
構造式 (3) - 4 :



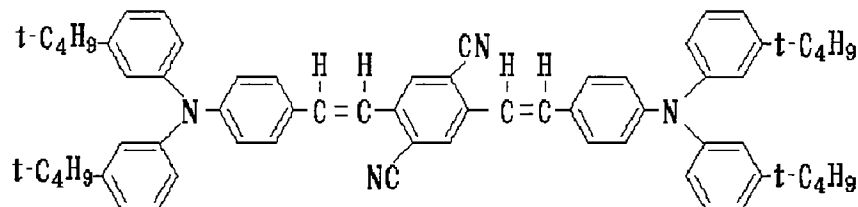
構造式 (3) - 5 :



構造式 (3) - 6 :



構造式 (3) - 7 :



【請求項6】 前記有機層が、正孔輸送層と電子輸送層とが積層された有機積層構造を有しており、前記正孔輸送層の形成材料として前記ジスチリル化合物が用いられている、請求項5に記載した有機電界発光素子。

【請求項7】 前記有機層が、正孔輸送層と電子輸送層とが順次積層された有機積層構造を有しており、前記電

られている、請求項5に記載した有機電界発光素子。

【請求項8】 前記有機層が、正孔輸送層と発光層と電子輸送層とが積層された有機積層構造を有しており、前記発光層の形成材料として前記ジスチリル化合物が用いられている、請求項5に記載した有機電界発光素子。

【発明の詳細な説明】

【発明の属する技術分野】本発明は、発光領域を有する有機層が陽極と陰極との間に設けられている有機電界発光素子（有機EL素子）に関するものである。

【0002】

【従来の技術】軽量で高効率のフラットパネルディスプレイが、例えばコンピュータやテレビジョンの画面表示用として盛んに研究、開発されている。

【0003】まず、ブラウン管（CRT）は、輝度が高く、色再現性が良いため、現在ディスプレイとして最も多く使われているが、嵩高く、重く、また消費電力も高いという問題がある。

【0004】また、軽量で高効率のフラットパネルディスプレイとして、アクティブマトリックス駆動などの液晶ディスプレイが商品化されている。しかしながら、液晶ディスプレイは、視野角が狭く、また、自発光でないため周囲が暗い環境下ではバックライトの消費電力が大きいことや、今後実用化が期待されている高精細度の高速ビデオ信号に対して十分な応答性能を有しない等の問題点がある。特に、大画面サイズのディスプレイを製造することは困難であり、そのコストが高い等の課題もある。

【0005】これに対する代替として、発光ダイオードを用いたディスプレイの可能性があるが、やはり製造コストが高く、また、1つの基板上に発光ダイオードのマトリックス構造を形成することが難しい等の問題があり、ブラウン管に取って代わる低価格のディスプレイ候補としては、実用化までの課題が大きい。

【0006】これらの諸課題を解決する可能性のあるフラットパネルディスプレイとして、最近、有機発光材料を用いた有機電界発光素子（有機EL素子）が注目されている。即ち、発光材料として有機化合物を用いることにより、自発光で、応答速度が高速であり、視野角依存性の無いフラットパネルディスプレイの実現が期待されている。

【0007】有機電界発光素子の構成は、透光性の正極と金属陰極との間に、電流の注入によって発光する発光材料を含む有機薄膜を形成したものである。C. W. Tang, S. A. VanSlyke等は Applied Physics Letters第51巻12号913～915頁（1987年）掲載の研究報告において、有機薄膜を正孔輸送性材料からなる薄膜と電子輸送性材料からなる薄膜との2層構造として、各々の電極から有機膜中に注入されたホールと電子が再結合することにより発光する素子構造を開発した（シングルヘテロ構造の有機EL素子）。

【0008】この素子構造では、正孔輸送材料または電子輸送材料のいずれかが発光材料を兼ねており、発光は発光材料の基底状態と励起状態のエネルギーギャップに対応した波長帯で起きる。このような2層構造とすることにより、大幅な駆動電圧の低減、発光効率の改善が行わ

【0009】その後、C. Adachi、S. Tokita、T. Tsutsui、S. Saito等の Japanese Journal of Applied Physics第27巻2号L269～L271頁（1988年）掲載の研究報告に記載されているように、正孔輸送材料、発光材料、電子輸送材料の3層構造（ダブルヘテロ構造の有機EL素子）が開発され、更に、C. W. Tang, S. A. VanSlyke, C. H. Chen等の Journal of Applied Physics 第65巻9号3610～3616頁（1989年）掲載の研究報告に記載されているように、電子輸送材料中に発光材料を含ませた素子構造などが開発された。これらの研究により、低電圧で、高輝度の発光の可能性が検証され、近年、研究開発が非常に活発に行われている。

【0010】発光材料に用いる有機化合物は、その多様性から、理論的には分子構造を変化させることによって発光色を任意に変えることができるという利点があると言える。従って、分子設計を施すことにより、フルカラーディスプレイに必要な色純度の良いR（赤）、G（緑）、B（青）の3色を揃えることは、無機物を用いた薄膜EL素子と比べて容易であると言える。

【0011】

【発明が解決しようとする課題】しかしながら、実際には有機電界発光素子においても、解決しなければならない問題がある。安定した高輝度の赤色発光素子の開発は難しく、現在報告されている電子輸送材料として、トリス（8-キノリノール）アルミニウム（以下、Alq₃と略称。）にDCM〔4-ジシアノメチレン-6-（p-ジメチルアミノスチリル）-2-メチル-4H-ピラン〕をドープした赤色発光の例においても、最高輝度、信頼性ともにディスプレイ材料としては満足の行くものではない。

【0012】また、T. Tsutsui, D. U. Kim が Inorganic and Organic electroluminescence 会議（1996、Berlin）で報告したBSB-BCNは、1000cd/m²以上の高い輝度を実現しているが、フルカラーに対応する赤色としての色度が完全なものとは言えない。

【0013】さらに高輝度で安定かつ色純度の高い赤色発光素子の実現が、望まれているのが現状である。

【0014】また、特開平7-188649号（特願平6-148798号）においては、特定のジスチリル化合物を有機電界発光材料とすることを提案しているが、目的の発光色が青色であり、赤色用ではない。

【0015】本発明の目的は、高輝度かつ安定な赤色発光を有する有機電界発光素子を提供することにある。

【0016】

【課題を解決するための手段】本発明は上記課題を解決するために鋭意検討した結果、発光材料として特定のジスチリル化合物を用いることによって、安定した、高輝度のフルカラーディスプレイ実現に極めて有用な信頼性

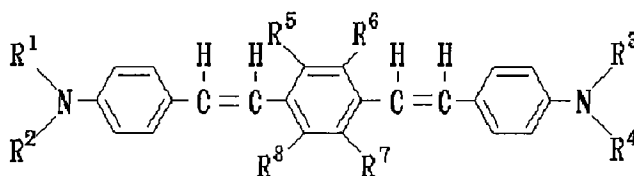
に到達したものである。

【0017】即ち、本発明は、発光領域を有する有機層が陽極と陰極との間に設けられ、電流の注入によって発光する有機物質を構成要素として含む有機電界発光素子

において、前記有機層に下記一般式（１）で表されるジスチリル化合物が有機発光材料として含まれていることを特徴とする、有機電界発光素子に係るものである。

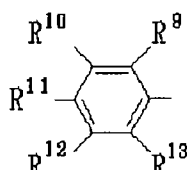
【化２】

一般式（１）：



〔但し、前記一般式（１）において、 R^1 、 R^2 、 R^3 及び R^4 は互いに同一の又は異なる基であって、下記一般式（２）で表されるアリール基であり

一般式（２）：



（但し、前記一般式（２）において、 R^9 、 R^{10} 、 R^{11} 、 R^{12} 及び R^{13} は互いに同一の又は異なる基であって、それらの少なくとも１つが飽和又は不飽和アルコキシ基、又はアルキル基（メチル基、ターシャリーブチル基が好ましく、その置換位置はメタ位が好ましい。）である。）、 R^5 、 R^6 、 R^7 及び R^8 は互いに同一の又は異なる基であって、それらの少なくとも１つがシア

ノ基、ニトロ基又はハロゲン原子（これにはF、Cl、Br、Iが挙げられる。）である。〕

【0018】上記一般式（１）のジスチリル化合物を発光材料に用いることによって、高輝度で安定な赤色発光が得られると共に、電氣的、熱的或いは化学的にも安定性に優れた素子を提供できる。

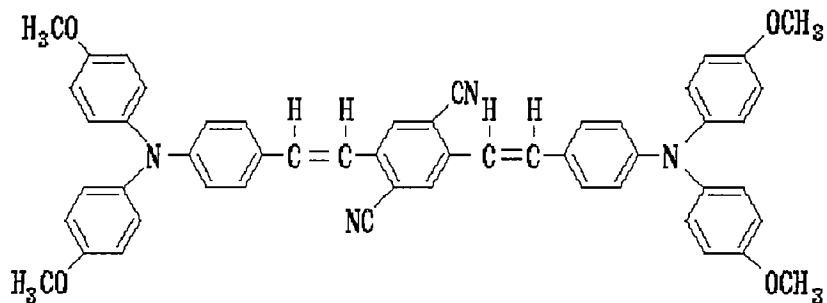
【0019】

【発明の実施の形態】本発明の有機電界発光素子に用いるジスチリル化合物について説明する。

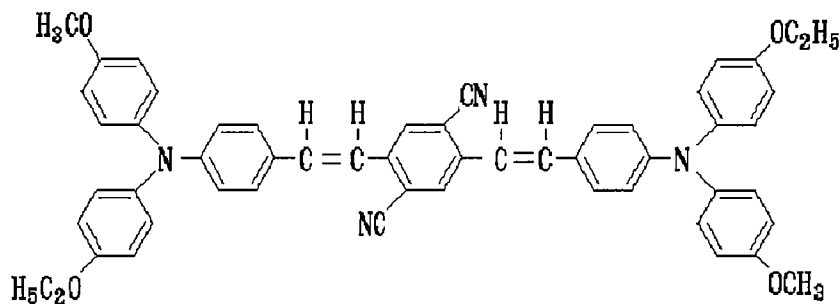
【0020】本発明の有機電界発光素子において、発光材料である一般式（１）で示されるジスチリル化合物は、例えば下記構造式（３）－１、（３）－２、（３）－３、（３）－４、（３）－５、（３）－６又は（３）－７のような分子構造の少なくとも１種が使用可能である。

【化３】

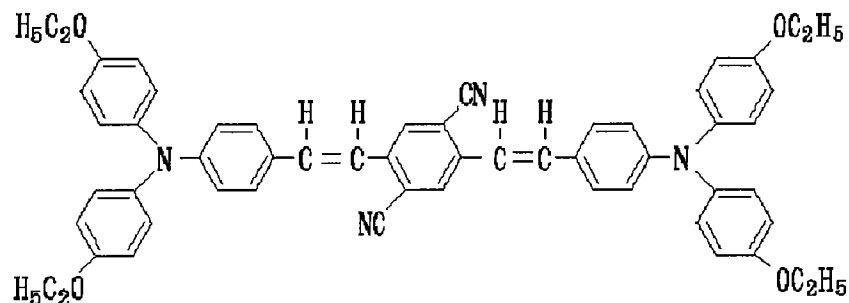
構造式（３）－１：



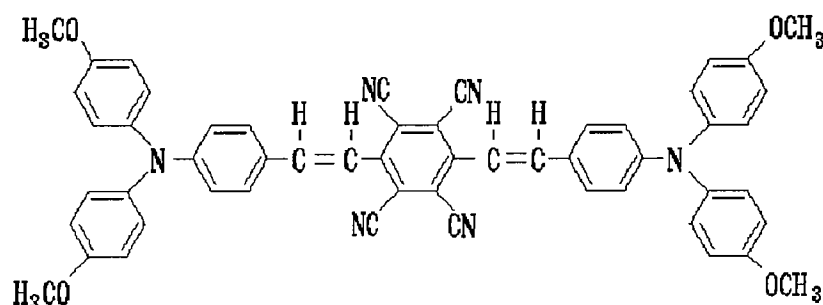
構造式（３）－２：



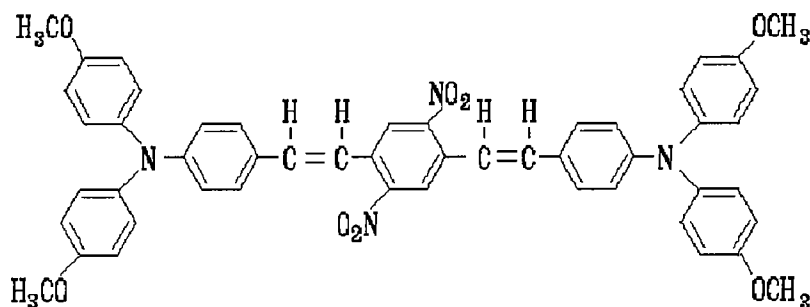
構造式 (3) - 3 :



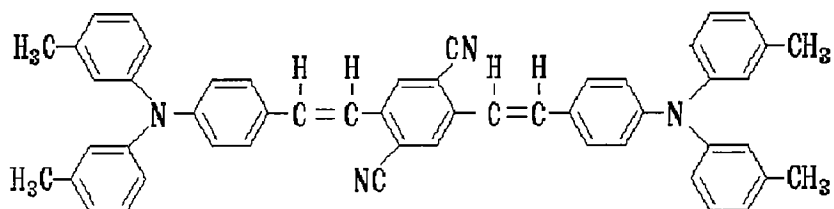
構造式 (3) - 4 :



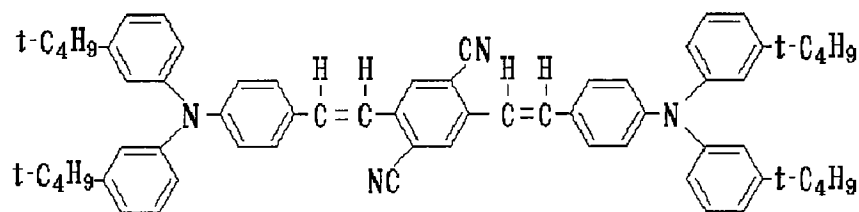
構造式 (3) - 5 :



構造式 (3) - 6 :



構造式 (3) - 7 :



【0021】図1～図4には、本発明に基づく有機電界発光素子の例をそれぞれ示す。

【0022】図1は陰極3を発光光20が透過する透過型有機電界発光素子Aであって、発光20は保護層4の側からも観測できる。図2は陰極3での反射光も発光光20として得る反射型有機電界発光素子Bを示す。

めの基板であり、ガラス、プラスチック及び他の適宜の材料を用いることができる。また、有機電界発光素子を他の表示素子と組み合わせて用いる場合には、基板を共用することもできる。2は透明電極（陽極）であり、ITO (Indium tin oxide)、 SnO_2 等を使用できる。

スチリル化合物を発光材料として含有している。この発光層について、有機電界発光20を得る層構成としては、従来公知の種々の構成を用いることができる。後述するように、例えば、正孔輸送層と電子輸送層のいずれかを構成する材料が発光性を有する場合、これらの薄膜を積層した構造を使用できる。更に本発明の目的を満たす範囲で電荷輸送性能を上げるために、正孔輸送層と電子輸送層のいずれか若しくは両方が、複数種の材料の薄膜を積層した構造、または、複数種の材料を混合した組成からなる薄膜を使用するのを妨げない。また、発光性能を上げるために、少なくとも1種以上の蛍光性の材料を用いて、この薄膜を正孔輸送層と電子輸送層の間に挟持した構造、更に少なくとも1種以上の蛍光性の材料を正孔輸送層若しくは電子輸送層、またはこれらの両方に含ませた構造を使用しても良い。これらの場合には、発光効率を改善するために、正孔または電子の輸送を制御するための薄膜をその層構成に含ませることも可能である。

【0025】上記の構造式(3)で例示したジスチリル化合物は、電子輸送性能と正孔輸送性能の両方を持つため、素子構成中、電子輸送層を兼ねた発光層としても、或いは正孔輸送層を兼ねた発光層としても用いることが可能である。また、このジスチリル化合物を発光層として、電子輸送層と正孔輸送層とで挟み込んだ構成とすることも可能である。

【0026】なお、図1及び図2中、3は陰極であり、電極材料としては、Li、Mg、Ca等の活性な金属とAg、Al、In等の金属との合金、或いはこれらを積層した構造を使用できる。透過型の有機電界発光素子においては、陰極の厚さを調節することにより、用途に合った光透過率を得ることができる。また、図4は封止・保護層であり、有機電界発光素子全体を覆う構造とすることにより、その効果が上がる。気密性が保たれば、適宜の材料を使用することができる。また、8は電流注入用の駆動電源である。

【0027】本発明に基づく有機電界発光素子において、有機層が、正孔輸送層と電子輸送層とが積層された有機積層構造(シングルヘテロ構造)を有しており、正孔輸送層又は電子輸送層の形成材料として前記ジスチリル化合物が用いられてよい。或いは、有機層が、正孔輸送層と発光層と電子輸送層とが順次積層された有機積層構造(ダブルヘテロ構造)を有しており、発光層の形成材料として前記ジスチリル化合物が用いられてよい。

【0028】このような有機積層構造を有する有機電界発光素子の例を示すと、図3は、透光性の基板1上に、透光性の陽極2と、正孔輸送層6と電子輸送層7とからなる有機層5aと、陰極3とが順次積層された積層構造を有し、この積層構造が保護膜4によって封止されてなる、シングルヘテロ構造の有機電界発光素子Cである。

の場合には、正孔輸送層6と電子輸送層7の界面から所定波長の発光20を発生する。これらの発光は基板1側から観測される。

【0030】また、図4は、透光性の基板1上に、透光性の陽極2と、正孔輸送層10と発光層11と電子輸送層12とからなる有機層5bと、陰極3とが順次積層された積層構造を有し、この積層構造が保護膜4によって封止されてなる、ダブルヘテロ構造の有機電界発光素子Dである。

【0031】図4に示した有機電界発光素子においては、陽極2と陰極3の間に直流電圧を印加することにより、陽極2から注入された正孔が正孔輸送層10を経て、また陰極3から注入された電子が電子輸送層12を経て、それぞれ発光層11に到達する。この結果、発光層11においては電子/正孔の再結合が生じて一重項励起子が生成し、この一重項励起子から所定波長の発光を発生する。

【0032】上述した各有機電界発光素子C、Dにおいて、基板1は、例えば、ガラス、プラスチック等の光透過性の材料を適宜用いることができる。また、他の表示素子と組み合わせて用いる場合や、図3及び図4に示した積層構造をマトリックス状に配置する場合等は、この基板を共用としてよい。また、素子C、Dはいずれも、透過型、反射型のいずれの構造も採りうる。

【0033】また、陽極2は、透明電極であり、ITO(indium tin oxide)や SnO_2 等が使用できる。この陽極2と正孔輸送層6(又は正孔輸送層10)との間には、電荷の注入効率を改善する目的で、有機物若しくは有機金属化合物からなる薄膜を設けてもよい。なお、保護膜4が金属等の導電性材料で形成されている場合は、陽極2の側面に絶縁膜が設けられていてもよい。

【0034】また、有機電界発光素子Cにおける有機層5aは、正孔輸送層6と電子輸送層7とが積層された有機層であり、これらのいずれか又は双方に上記したジスチリル化合物が含有され、発光性の正孔輸送層6又は電子輸送層7としてよい。有機電界発光素子Dにおける有機層5bは、正孔輸送層10と上記したジスチリル化合物を含有する発光層11と電子輸送層12とが積層された有機層であるが、その他、種々の積層構造を取ることができる。例えば、正孔輸送層と電子輸送層のいずれか若しくは両方が発光性を有していてもよい。

【0035】また、特に、正孔輸送層6又は電子輸送層7や発光層11が本発明のジスチリル化合物からなる層であることが望ましいが、これらの層を前記ジスチリル化合物のみで形成してもよく、或いは、前記ジスチリル化合物と他の正孔又は電子輸送材料(例えば、芳香族アミン類やピラゾリン類等)との共蒸着によって形成してもよい。さらに、正孔輸送層において、正孔輸送性能を向上させるために、複数種の正孔輸送材料を積層した正

【0036】また、有機電界発光素子Cにおいて、発光層は電子輸送性発光層7であってよいが、電源8から印加される電圧によっては、正孔輸送層6やその界面で発光される場合がある。同様に、有機電界発光素子Dにおいて、発光層は層11以外に、電子輸送層12であってよく、正孔輸送層10であってよい。発光性能を向上させるために、少なくとも1種の蛍光性材料を用いた発光層11を正孔輸送層と電子輸送層との間に挟持させた構造であるのがよい。または、この蛍光性材料を正孔輸送層又は電子輸送層、或いはこれら両層に含有させた構造を構成してよい。このような場合、発光効率を改善するために、正孔又は電子の輸送を制御するための薄膜（ホールブロッキング層やエキシトン生成層など）をその層構成に含ませることも可能である。

【0037】また、陰極3に用いる材料としては、Li、Mg、Ca等の活性な金属とAg、Al、In等の金属との合金を使用でき、これらの金属層が積層した構造であってもよい。なお、陰極の厚みや材質を適宜選択することによって、用途に見合った有機電界発光素子を作製できる。

【0038】また、保護膜4は、封止膜として作用するものであり、有機電界発光素子全体を覆う構造とすることで、電荷注入効率や発光効率を向上できる。なお、その気密性が保たれば、アルミニウム、金、クロム等の単金属又は合金など、適宜その材料を選択できる。

【0039】上記した各有機電界発光素子に印加する電流は通常、直流であるが、パルス電流や交流を用いてもよい。電流値、電圧値は、素子破壊しない範囲内であれば特に制限はないが、有機電界発光素子の消費電力や寿命を考慮すると、なるべく小さい電気エネルギーで効率良く発光させることが望ましい。

【0040】次に、図5は、本発明の有機電界発光素子を用いた平面ディスプレイの構成例である。図示の如く、例えばフルカラーディスプレイの場合は、赤

(R)、緑(G)及び青(B)の3原色を発光可能な有機層5(5a、5b)が、陰極3と陽極2との間に配されている。陰極3及び陽極2は、互いに交差するストライプ状に設けることができ、輝度信号回路14及びシフトレジスタ内蔵の制御回路15により選択されて、それぞれに信号電圧が印加され、これによって、選択された陰極3及び陽極2が交差する位置(画素)の有機層が発光するように構成される。

【0041】即ち、図5は例えば8×3RGB単純マトリックスであって、正孔輸送層と、発光層および電子輸送層のいずれか少なくとも一方とからなる積層体5を陰極3と陽極2の間に配置したものである(図3又は図4参照)。陰極と陽極は、ともにストライプ状にパターンニングするとともに、互いにマトリクス状に直交させ、シフトレジスタ内蔵の制御回路15および14により時系列的に信号電圧を印加し、その交叉位置で発光するように構成されたものである。かかる構成のEL素子は、文字・記号等のディスプレイとしては勿論、画像再生装置としても使用できる。また陰極3と陽極2のストライプ状パターンを赤(R)、緑(G)、青(B)の各色毎に配し、マルチカラーあるいはフルカラーの全固体型フラットパネルディスプレイを構成することが可能となる。

【0042】

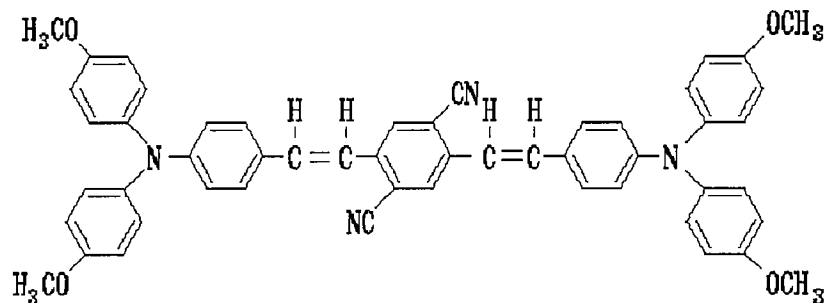
【実施例】以下、本発明を実施例について具体的に説明するが、本発明は以下の実施例に限定されるものではない。

【0043】実施例1

本実施例は、一般式(1)の上記ジスチリル化合物のうち、R¹、R²、R³、R⁴に3-メトキシフェニル基を、R⁶、R⁸にシアノ基を持った下記構造式(3)-1の化合物を正孔輸送性発光材料として用い、シングルヘテロ構造の有機電界発光素子を作製した例である。

【化4】

構造式(3)-1:



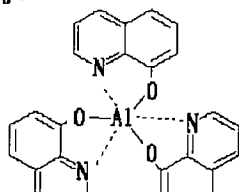
【0044】まず、真空蒸着装置中に、100nmの厚さのITOからなる陽極が一表面に形成された30mm×30mmのガラス基板をセッティングした。蒸着マスクとして、複数の2.0mm×2.0mmの単位開口を有する金属マスクを基板に近接して配置し、真空蒸着法により10⁻⁴Pa以下の真空下で上記構造式(3)-1

層)として成膜した。蒸着レートは0.1nm/秒とした。

【0045】さらに、電子輸送材料として下記構造式のAlq₃(トリス(8-キノリノール)アルミニウム)を正孔輸送層に接して蒸着した。Alq₃からなるこの電子輸送層の膜厚も例えば50nmとし、蒸着レートは

【化5】

Alq₃ :



【0046】陰極材料としてはMgとAgの積層膜を採用し、これも蒸着により、蒸着レート1nm/秒として例えば50nm(Mg膜)および150nm(Ag膜)の厚さに形成し、実施例1による図3に示した如き有機電界発光素子を作製した。

【0047】このように作製した実施例1の有機電界発光素子に、窒素雰囲気下で順バイアス直流電圧を加えて発光特性を評価した。発光色は赤色であり、分光測定を行った結果、図6に示すように、650nmに発光ピークを有するスペクトルを得た。分光測定は、大塚電子社製のフォトダイオードアレイを検出器とした分光器を用いた。また、電圧-輝度測定を行ったところ、図8に示すように、9.5Vで1200cd/m²の輝度が得られた。

【0048】この有機電界発光素子を作製後、窒素雰囲気下に1カ月間放置したが、素子劣化は観察されなかった。また、初期輝度200cd/m²で電流値を一定に通電して連続発光し、強制劣化させた際、輝度が半減するまで1000時間であった。

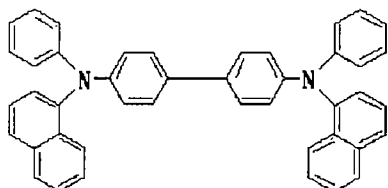
【0049】実施例2

本実施例は、一般式(1)の上記ジスチリル化合物のうち、R¹、R²、R³、R⁴に3-メトキシフェニル基を、R⁶、R⁸にシアノ基を持った上記構造式(3)-1の化合物を電子輸送性発光材料として用い、シングルヘテロ構造の有機電界発光素子を作製した例である。

【0050】まず、真空蒸着装置中に、100nmの厚さのITOからなる陽極が一表面に形成された30mm×30mmのガラス基板をセッティングした。蒸着マスクとして、複数の2.0mm×2.0mmの単位開口を有する金属マスクを基板に近接して配置し、真空蒸着法により10⁻⁴Pa以下の真空下で、下記構造式のα-NPD(α-ナフチルフェニルジアミン)を例えば50nmの厚さに正孔輸送層として成膜した。蒸着レートは0.1nm/秒とした。

【化6】

α-NPD :



(3)-1の化合物を正孔輸送層に接して蒸着した。上記構造式(3)-1の化合物からなる電子輸送層(兼発光層)の膜厚も例えば50nmとし、蒸着レートは0.2nm/秒とした。

【0052】陰極材料としてはMgとAgの積層膜を採用し、これも蒸着により、蒸着レート1nm/秒として例えば50nm(Mg膜)および150nm(Ag膜)の厚さに形成し、実施例2による図3に示した如き有機電界発光素子を作製した。

【0053】このように作製した実施例2の有機電界発光素子に、窒素雰囲気下で順バイアス直流電圧を加えて発光特性を評価した。発光色は赤色であり、実施例1と同様に分光測定を行った結果、図7に示すように、650nmに発光ピークを有するスペクトルを得た。また、電圧-輝度測定を行ったところ、図9に示すように、10.5Vで600cd/m²の輝度が得られた。

【0054】この有機電界発光素子を作製後、窒素雰囲気下に1カ月間放置したが、素子劣化は観察されなかった。また、初期輝度200cd/m²で電流値を一定に通電して連続発光し、強制劣化させた際、輝度が半減するまで700時間であった。

【0055】実施例3

本実施例は、一般式(1)の上記ジスチリル化合物のうち、R¹、R²、R³、R⁴に3-メトキシフェニル基を、R⁶、R⁸にシアノ基を持った上記構造式(3)-1の化合物を発光材料として用い、ダブルヘテロ層構造の有機電界発光素子を作製した例である。

【0056】まず、真空蒸着装置中に、100nmの厚さのITOからなる陽極が一表面に形成された30mm×30mmのガラス基板をセッティングした。蒸着マスクとして、複数の2.0mm×2.0mmの単位開口を有する金属マスクを基板に近接して配置し、真空蒸着法により10⁻⁴Pa以下の真空下で、上記構造式のα-NPDを例えば30nmの厚さに正孔輸送層として成膜した。蒸着レートは0.2nm/秒とした。

【0057】さらに、発光材料として上記構造式(3)-1の化合物を正孔輸送層に接して蒸着した。上記構造式(3)-1の化合物からなる発光層の膜厚も例えば30nmとし、蒸着レートは0.2nm/秒とした。

【0058】さらに、電子輸送材料として上記構造式のAlq₃を発光層に接して蒸着した。Alq₃の膜厚を例えば30nmとし、蒸着レートは、0.2nm/秒とした。

【0059】陰極材料としてはMgとAgの積層膜を採用し、これも蒸着により、蒸着レート1nm/秒として例えば50nm(Mg膜)および150nm(Ag膜)の厚さに形成し、実施例3による図4に示した如き有機電界発光素子を作製した。

【0060】このように作製した実施例3の有機電界発

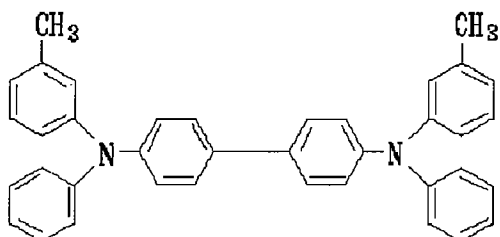
発光特性を評価した。発光色は赤色であり、分光測定を行った結果、650nmに発光ピークを有するスペクトルを得た。電圧－輝度測定を行ったところ、8.5Vで1800cd/m²の輝度が得られた。

【0061】この有機電界発光素子を作製後、窒素雰囲気下に1カ月間放置したが、素子劣化は観察されなかった。また、初期輝度200cd/m²で電流値を一定に通電して連続発光し、強制劣化させた際、輝度が半減するまで1500時間であった。

【0062】実施例4

正孔輸送性材料として α -NPDに替えて下記構造式のTPD（トリフェニルジアミン誘導体）を用いた他は層構成、成膜法とも実施例2に準拠して、有機電界発光素子を作製した。

TPD：



【0063】本実施例の有機電界発光素子も実施例2と同様の赤色の発光を呈した。分光測定の結果、スペクトルは実施例2の有機電界発光素子のスペクトルと一致した。

【0064】

【発明の作用効果】本発明の有機電界発光素子によれば、発光領域を有する有機層が陽極と陰極との間に設けられている有機電界発光素子において、前記有機層に前

記一般式（1）で表されるジスチリル化合物が含まれているので、高輝度で安定な赤色発光を有する有機電界発光素子を提供することが可能となる。

【図面の簡単な説明】

【図1】本発明に基づく有機電界発光素子の要部概略断面図である。

【図2】同、有機電界発光素子の他の要部概略断面図である。

【図3】同、有機電界発光素子の他の要部概略断面図である。

【図4】同、有機電界発光素子の他の要部概略断面図である。

【図5】同、有機電界発光素子を用いたフルカラーの平面ディスプレイの構成図である。

【図6】本発明の実施例1による有機電界発光素子の発光スペクトル図である。

【図7】同、実施例2による有機電界発光素子の発光スペクトル図である。

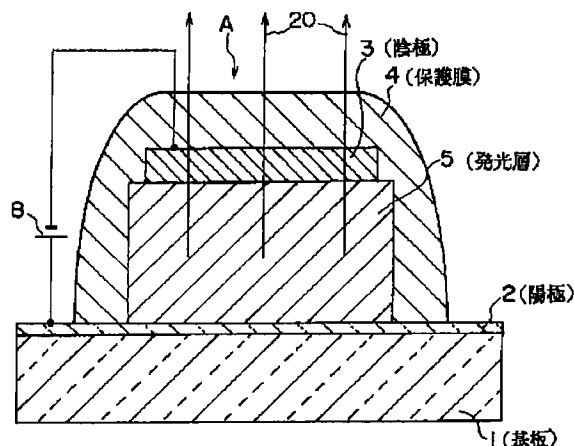
【図8】同、実施例1による有機電界発光素子の電圧－輝度特性図である。

【図9】同、実施例2による有機電界発光素子の電圧－輝度特性図である。

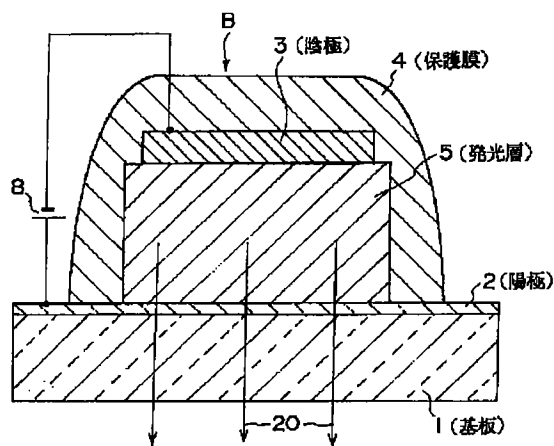
【符号の説明】

1…基板、2…透明電極（陽極）、3…陰極、4…保護膜、5、5a、5b…有機層、6…正孔輸送層、7…電子輸送層、8…電源、10…正孔輸送層、11…発光層、12…電子輸送層、14…輝度信号回路、15…制御回路、20…発光光、A、B、C、D…有機電界発光素子

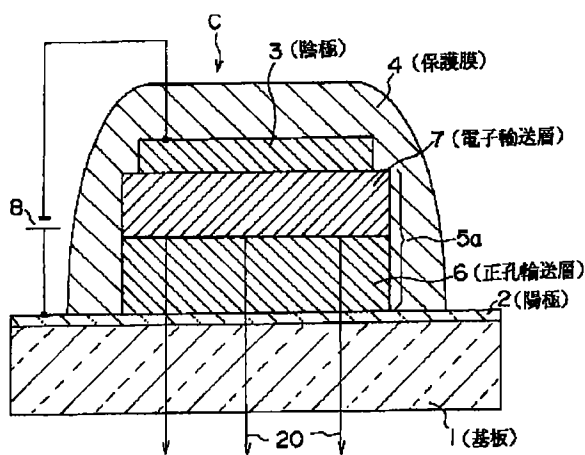
【図1】



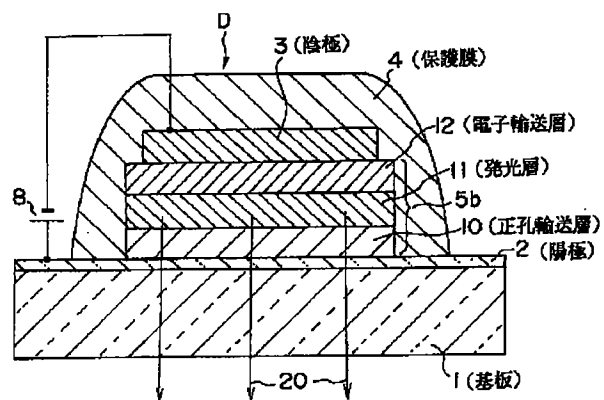
【図2】



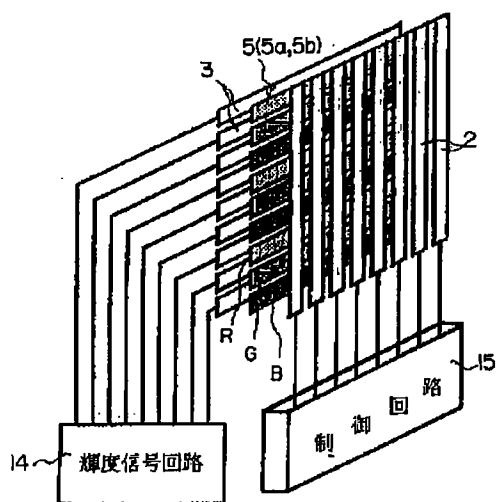
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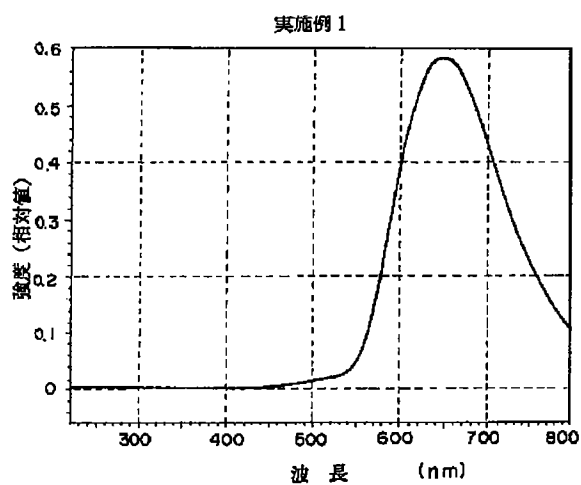
【図4】



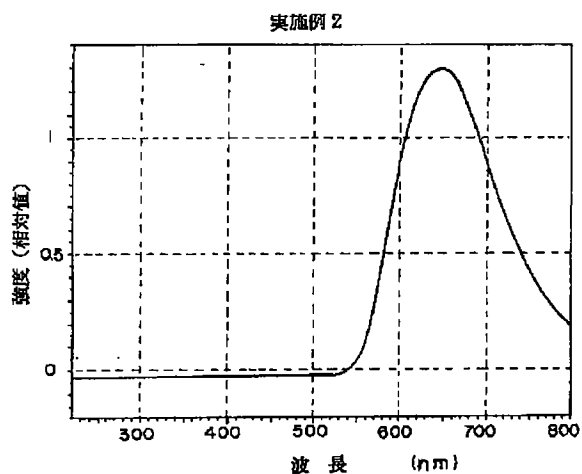
【図5】



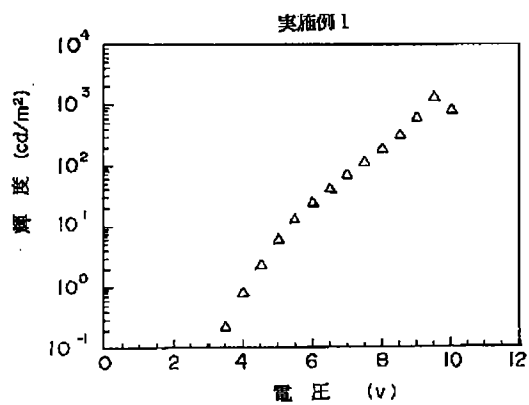
【図6】



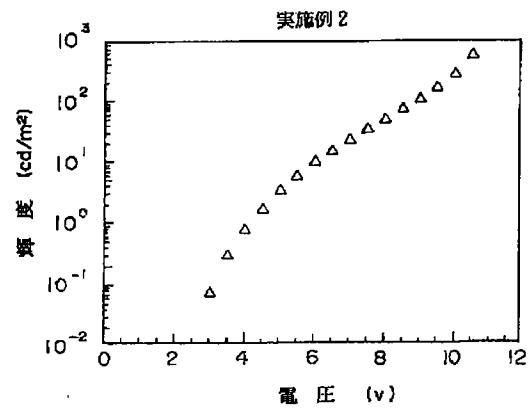
【図7】



【図8】



【図9】



PATENT ABSTRACTS OF JAPAN

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(22)Date of filing : 18.05.1998 (72)Inventor : ISHIBASHI TADASHI

ICHIMURA MARI

(54) ORGANIC ELECTROLUMINESCENT ELEMENT

(57)Abstract:

PROBLEM TO BE SOLVED: To provide an electroluminescent element having highly bright and stable red luminescence by using a distyryl compound as a luminescent material.

SOLUTION: An organic layer contains a distyryl compound represented by formula I. In the formula I, R1, R2, R3, and R4 are the same or a different group, and an aryl group represented by formula II, R5, R6, R7, and R8 are the same or a different group, and at least one of them is an amino group, a nitro group, or a halogen atom. R9-R13 in formula II are the same or a different group, and at least one of them is a saturated or unsaturated alkoxy group, or an alkyl group. A compound represented by formula III, for example, of the distyryl compound can be used as a luminescent layer also acting as an electron transport layer, or a luminescent layer also acting as a hole transport layer, and can be constituted as a luminescent layer interposed between the electron transport layer and the

hole transport layer.

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[Kind of final disposal of application other than the examiner's decision of rejection or application converted registration]

[Date of final disposal for application]

[Patent number] 3852518

[Date of registration] 15.09.2006

[Number of appeal against examiner's decision of rejection]

[Date of requesting appeal against examiner's decision of rejection]

[Date of extinction of right]

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3.In the drawings, any words are not translated.

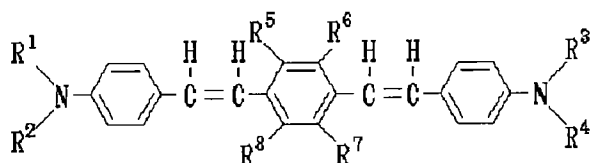
CLAIMS

[Claim(s)]

[Claim 1] Organic electroluminescence devices characterized by containing the JISUCHIRIRU compound expressed with said organic layer by the following general formula (1) as an organic luminescent material in the organic electroluminescence devices by which the organic layer which has a luminescence field is prepared between an anode plate and cathode.

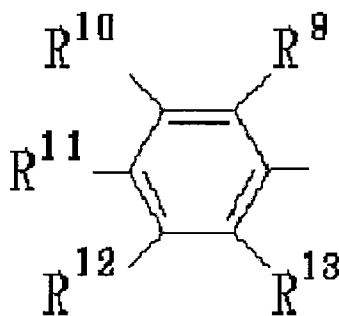
[Formula 1]

一般式 (1) :



[-- however, said general formula (1) -- setting -- R1, R2, R3, and R4 -- mutual --
identitas -- or the aryl group which is a different radical and is expressed with the

一般式 (2) :



following general formula (2) -- it is

(However, in said general formula (2), R⁹, and R¹⁰, R¹¹, R¹² and R¹³ are radicals which are identical mutually or are different.) those at least one is saturation, a partial saturation alkoxy group, or an alkyl group. R⁵, R⁶, and R⁷ And R⁸ mutual -- identical -- or it is a different radical and those at least one is a cyano group, a nitro group, or a halogen atom.]

[Claim 2] Organic electroluminescence devices for which said organic layer has the organic laminated structure to which the laminating of an electron hole transportation layer and the electron transport layer was carried out, and said JISUCHIRIRU compound is used as a formation ingredient of said electron hole transportation layer and which were indicated to claim 1.

[Claim 3] Organic electroluminescence devices for which said organic layer has the organic laminated structure to which the laminating of an electron hole transportation layer and the electron transport layer was carried out one by one, and said JISUCHIRIRU compound is used as a formation ingredient of said

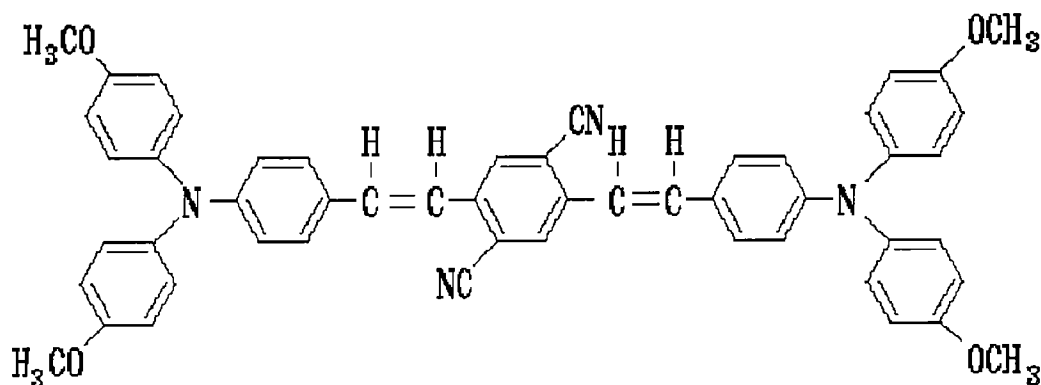
electron transport layer and which were indicated to claim 1.

[Claim 4] Organic electroluminescence devices for which said organic layer has the organic laminated structure to which the laminating of an electron hole transportation layer, a luminous layer, and the electron transport layer was carried out, and said JISUCHIRIRU compound is used as a formation ingredient of said luminous layer and which were indicated to claim 1.

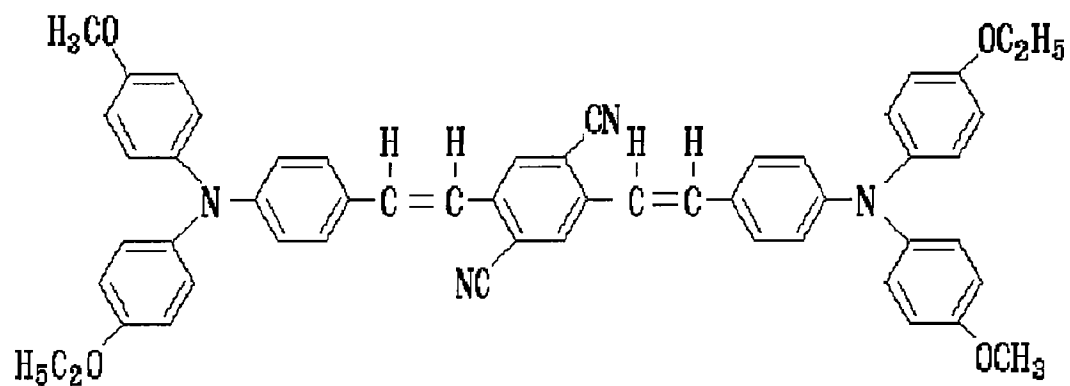
[Claim 5] Organic electroluminescence devices characterized by containing at least one sort of a JISUCHIRIRU compound expressed with said organic layer following structure-expression (3)-1, (3)-2, (3)-3, (3)-4, (3)-5, (3)-6, or (3)-7 as an organic luminescent material in the organic electroluminescence devices by which the organic layer which has a luminescence field is prepared between an anode plate and cathode.

[Formula 2]

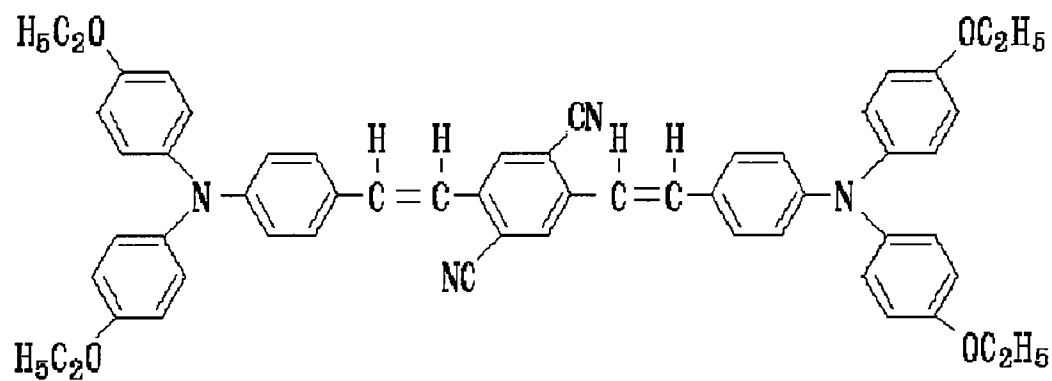
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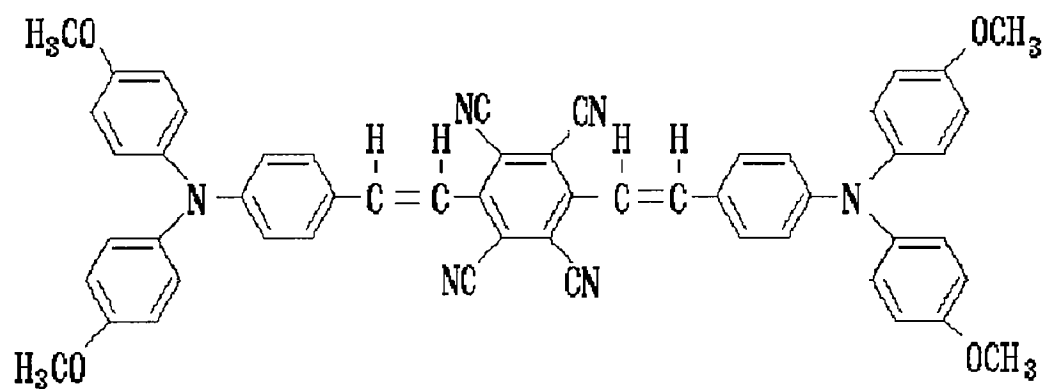
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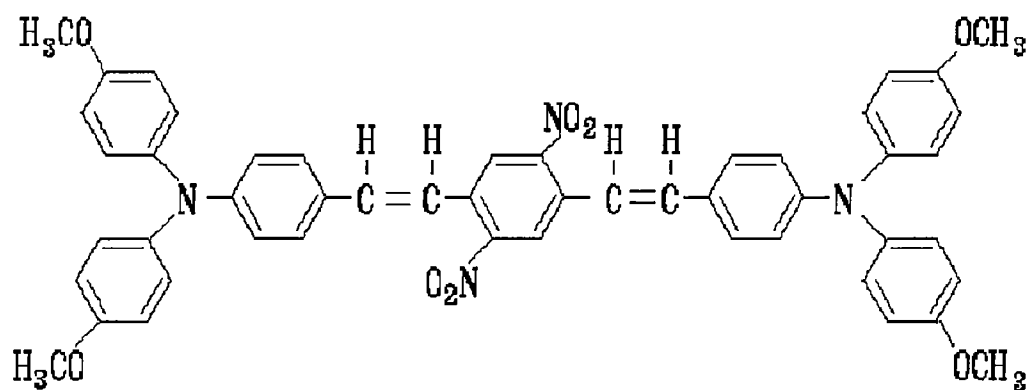
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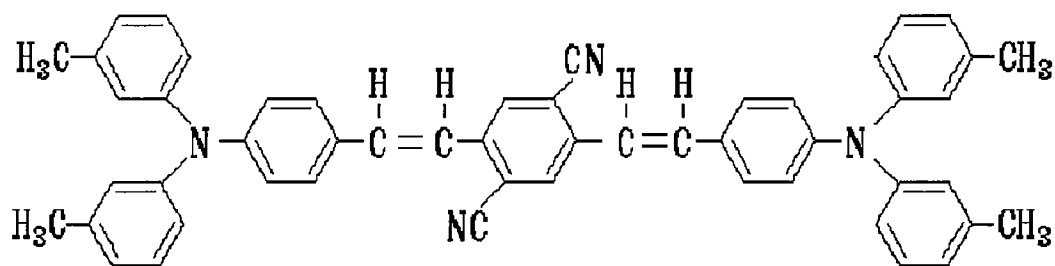
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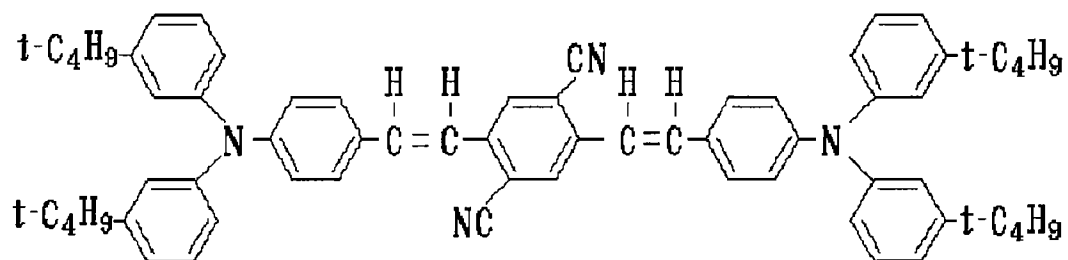
構造式 (3) - 5 :



構造式 (3) - 6 :



構造式 (3) - 7 :



[Claim 6] Organic electroluminescence devices for which said organic layer has the organic laminated structure to which the laminating of an electron hole transportation layer and the electron transport layer was carried out, and said

JISUCHIRIRU compound is used as a formation ingredient of said electron hole transportation layer and which were indicated to claim 5.

[Claim 7] Organic electroluminescence devices for which said organic layer has the organic laminated structure to which the laminating of an electron hole transportation layer and the electron transport layer was carried out one by one, and said JISUCHIRIRU compound is used as a formation ingredient of said electron transport layer and which were indicated to claim 5.

[Claim 8] Organic electroluminescence devices for which said organic layer has the organic laminated structure to which the laminating of an electron hole transportation layer, a luminous layer, and the electron transport layer was carried out, and said JISUCHIRIRU compound is used as a formation ingredient of said luminous layer and which were indicated to claim 5.

DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the organic electroluminescence devices (organic EL device) by which the organic layer which has a

luminescence field is prepared between an anode plate and cathode.

[0002]

[Description of the Prior Art] The lightweight and efficient flat-panel display is briskly studied and developed as an object for the screen display of a computer or television.

[0003] first -- although brightness of the Braun tube (CRT) is high, and it is used most mostly as a current display since color reproduction nature is good -- ** -- it is high and power consumption also has heavily the problem of being high.

[0004] Moreover, liquid crystal displays, such as an active-matrix drive, are commercialized as a lightweight and efficient flat-panel display. However, the angle of visibility of a liquid crystal display is narrow, and since it is not spontaneous light, it has the trouble of not having sufficient response engine performance to that the power consumption of a back light is large, and the high-speed video signal of a high definition with which utilization will be expected from now on under an environment dark in a perimeter. Technical problems, like especially the thing for which the display of big screen size is manufactured is difficult, and the cost is high also occur.

[0005] Although there is possibility of the display using the light emitting diode as an alternative over this, too, a manufacturing cost is high and the technical problem to utilization is large as a display candidate of a low price who there are

problems, like it is difficult to form the matrix structure of a light emitting diode on the substrate whose number is one, and replaces the Braun tube.

[0006] Organic electroluminescence devices (organic EL device) using an organic luminescent material as a flat-panel display which may solve many of these technical problems attract attention recently. That is, by using an organic compound as a luminescent material, a speed of response is high-speed with spontaneous light, and implementation of a flat-panel display without an angle-of-visibility dependency is expected.

[0007] The configuration of organic electroluminescence devices forms the organic thin film containing the luminescent material which emits light by impregnation of a current between the positive electrode of translucency, and metal cathode. C. W.Tang, S.A.VanSlyke, etc. In the research report of the 51st-volume No. 12 Applied Physics Letters 913-915-page (1987) printing The component structure which emits light when the hole and electron which were poured in into the organic film from each electrode recombine an organic thin film as two-layer structure of the thin film which consists of an electron hole transportability ingredient, and the thin film which consists of an electronic transportability ingredient was developed (organic EL device of single hetero structure).

[0008] With this component structure, either the electron hole transportation

ingredient or the electronic transportation ingredient serves as luminescent material, and luminescence occurs by the wavelength range corresponding to the energy gap of the ground state and excitation state of luminescent material. By considering as such two-layer structure, reduction of large driver voltage and an improvement of luminous efficiency were made.

[0009] Then, C.Adachi, S.Tokita, T.Tsutsui, S.Saito etc. As indicated by the research report of Japanese Journal of Applied Physics volume [27th] No. 2 L269-L 271-page (1988) printing The three-tiered structure (organic EL device of double hetero structure) of an electron hole transportation ingredient, luminescent material, and an electronic transportation ingredient is developed. Furthermore, C.W.Tang, S.A.VanSlyke, C.H.Chen, etc. Journal of Applied Physics As indicated by the research report of the 65th-volume No. 9 3610-3616-page (1989) printing The component structure where luminescent material was included in the electronic transportation ingredient etc. was developed. By these researches, by the low battery, the possibility of luminescence of high brightness is verified and researches and developments are done very actively in recent years.

[0010] It can be said that there is an organic compound used for luminescent material about the advantage that the luminescent color is theoretically changeable into arbitration by changing the molecular structure from the

versatility. Therefore, it can be said to be easy by performing a molecular design compared with the thin film EL element which used the inorganic substance to arrange three good colors of R (red), G (green), and B (blue) of color purity required for a full color display.

[0011]

[Problem(s) to be Solved by the Invention] However, also in organic electroluminescence devices, there is a problem which must be solved in fact. Development of the stable red light emitting device of high brightness is difficult, and the satisfaction as a display ingredient of the highest brightness and dependability does not go as an electronic transportation ingredient by which the current report is carried out in the example of red luminescence which doped DCM [a 4-dicyanomethylene-6-(p-dimethylaminostyryl)-2-methyl-4H-pyran] to tris (eight quinolinol) aluminum (the following, Alq3, and abbreviated name.).

[0012] Moreover, T.Tsutsui and D.U.Kim Inorganic and Organic electroluminescence BSB-BCN reported at the meeting (1996 Berlin) is 1000 cd/m². Although the above high brightness is realized, it cannot be said to be what has a perfect chromaticity as red which corresponds in full color.

[0013] The present condition is that implementation of stability and the high red light emitting device of color purity is furthermore desired by high brightness.

[0014] Moreover, in JP,7-188649,A (Japanese Patent Application No. No.

148798 [six to]), although it has proposed using a specific JISUCHIRIRU compound as an organic electroluminescence ingredient, the target luminescent color is blue and it is not an object for red.

[0015] The purpose of this invention is to offer the organic electroluminescence devices which have high brightness and stable red luminescence.

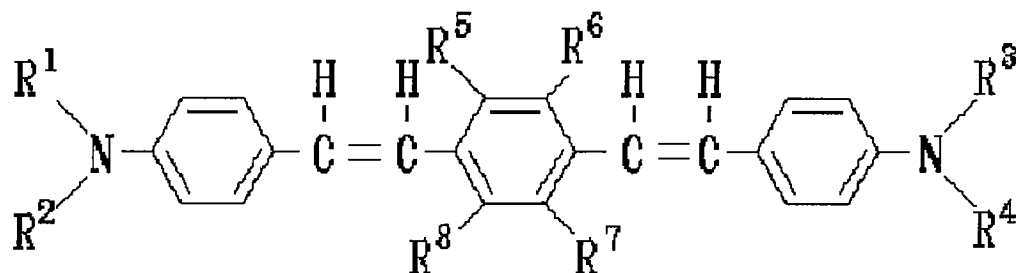
[0016]

[Means for Solving the Problem] In order that this invention may solve the above-mentioned technical problem, as a result of inquiring wholeheartedly, a header and this invention are reached [that the stable full color display implementation of high brightness can be provided with a red light emitting device with very useful high dependability, and] by using a specific JISUCHIRIRU compound as a luminescent material.

[0017] That is, this invention relates to the organic electroluminescence devices which the organic layer which has a luminescence field is prepared between an anode plate and cathode, and are characterized by containing the JISUCHIRIRU compound expressed with said organic layer by the following general formula (1) as an organic luminescent material in the organic electroluminescence devices which contain as a component the organic substance which emits light by impregnation of a current.

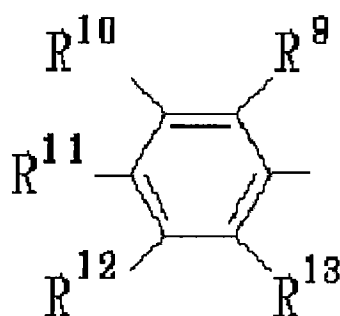
[Formula 2]

一般式 (1) :



[-- however, said general formula (1) -- setting -- R1, R2, R3, and R4 -- mutual -- identitas -- or the aryl group which is a different radical and is expressed with the

一般式 (2) :



following general formula (2) -- it is

(However, in said general formula (2), R9, and R10, R11, R12 and R13 are radicals which are identitas mutually or are different.) Those at least one is saturation, a partial saturation alkoxyl group, or an alkyl group (a methyl group and tertiary butyl are desirable and the permutation location has the desirable meta position.). it is . R5, R6, and R7 are radicals which are identitas mutually or are different, and those at least one is a cyano group, a nitro group, or a halogen atom (F, Cl, Br, and I are mentioned to this.). And R8 it is .]

[0018] While stable red luminescence is obtained by high brightness by using the JISUCHIRIRU compound of the above-mentioned general formula (1) for luminescent material, the component which was chemically [electrically, thermally, or] excellent in stability can be offered.

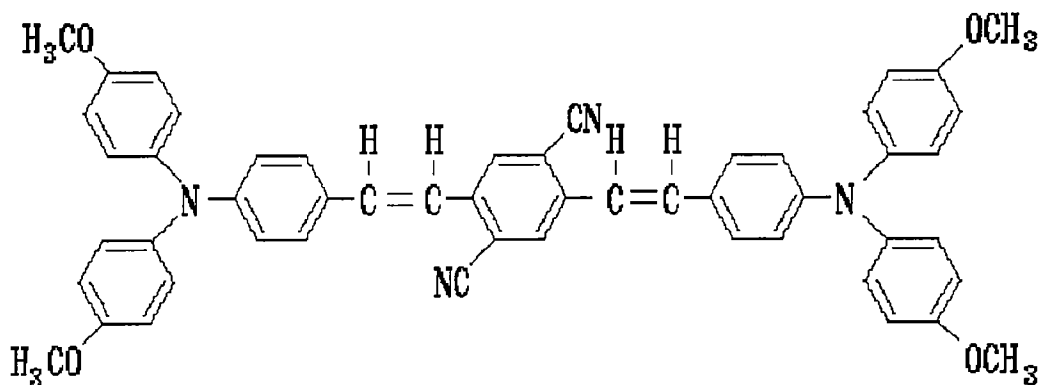
[0019]

[Embodiment of the Invention] The JISUCHIRIRU compound used for the organic electroluminescence devices of this invention is explained.

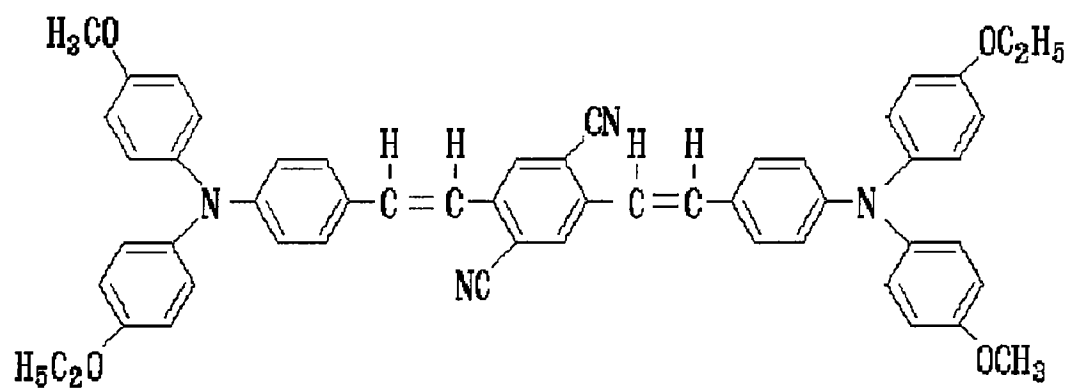
[0020] In the organic electroluminescence devices of this invention, the JISUCHIRIRU compound shown by the general formula (1) which is luminescent material has at least one usable sort of the molecular structure like following structure-expression (3)-1, (3)-2, (3)-3, (3)-4, (3)-5, (3)-6, or (3)-7.

[Formula 3]

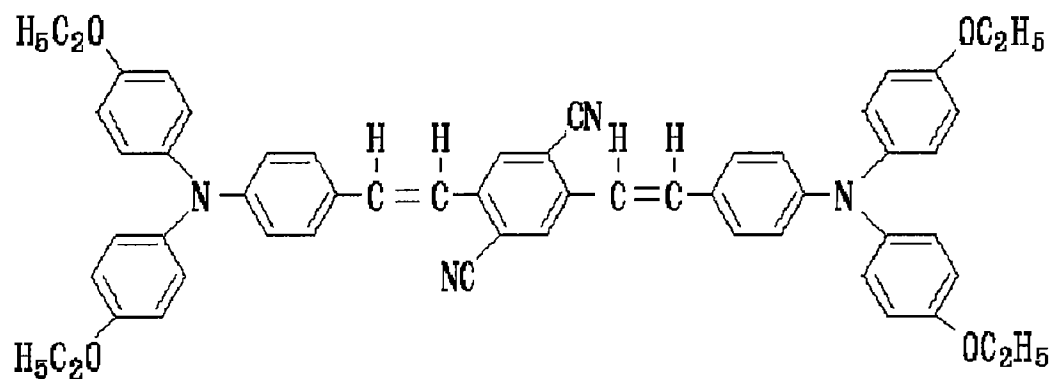
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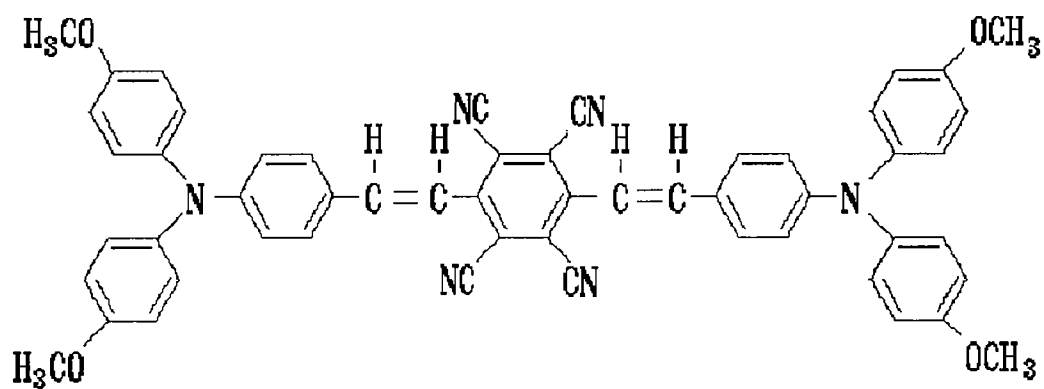
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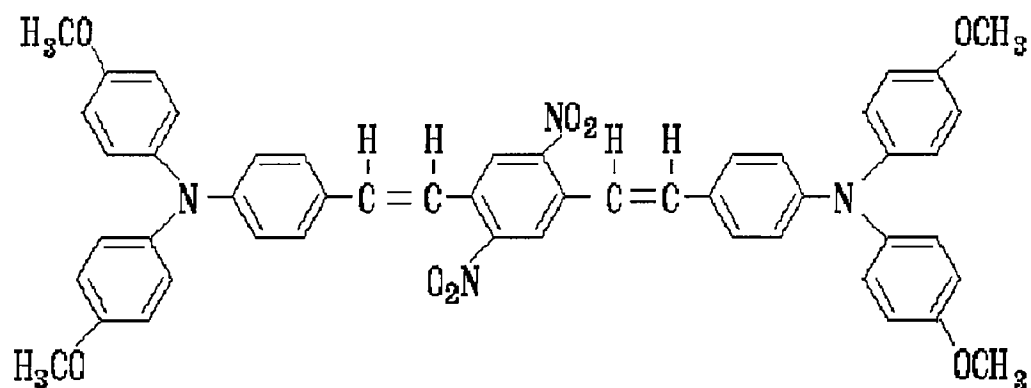
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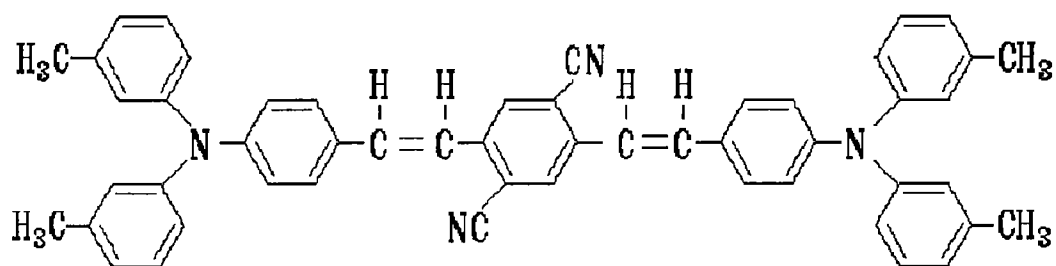
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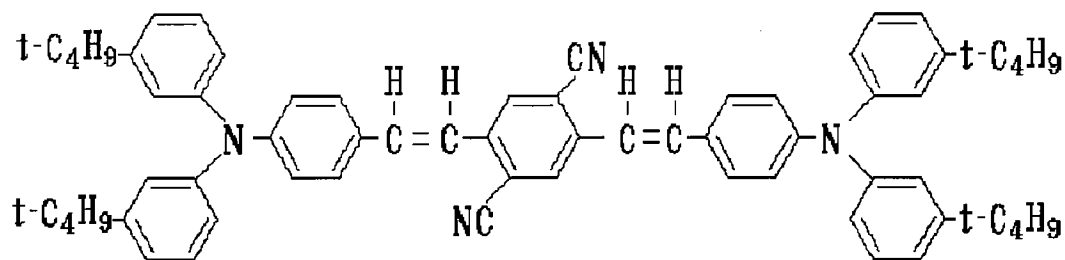
構造式 (3) - 5 :



構造式 (3) - 6 :



構造式 (3) - 7 :



[0021] The example of organic electroluminescence devices based on this invention is shown in drawing 1 - drawing 4 , respectively.

[0022] Drawing 1 is the transparency mold organic electroluminescence devices

A to which the luminescence light 20 penetrates cathode 3, and luminescence 20 can be observed also from a protective layer 4 side. Drawing 2 shows the reflective mold organic electroluminescence devices B which also obtain the reflected light in cathode 3 as a luminescence light 20.

[0023] Among drawing, one is a substrate for forming organic electroluminescence devices, and can use glass, plastics, and other proper ingredients. Moreover, a substrate can also be shared when using organic electroluminescence devices combining other display devices. 2 -- a transparent electrode (anode plate) -- it is -- ITO (Indium tin oxide) and SnO₂ etc. -- it can be used.

[0024] Moreover, 5 is an organic luminous layer and contains the above-mentioned JISUCHIRIRU compound as a luminescent material. About this luminous layer, well-known various configurations can be conventionally used as lamination which obtains organic electroluminescence 20. When the ingredient which constitutes an electron hole transportation layer or an electron transport layer has a luminescence so that it may mention later for example, the structure which carried out the laminating of these thin films can be used. Furthermore, in order to raise charge transportability ability in the range which fills the purpose of this invention, both an electron hole transportation layer, and both [either or] bar using the structure which carried out the laminating of the

thin film of two or more sorts of ingredients, or the thin film which consists of a presentation which mixed two or more sorts of ingredients. Moreover, in order to improve the luminescence engine performance, the ingredient of at least one or more sorts of fluorescence may be used, and the structure which pinched this thin film between the electron hole transportation layer and the electron transport layer, and the structure where the ingredient of at least one or more sorts of fluorescence was further included in an electron hole transportation layer, electron transport layers, or these both may be used. In order to improve luminous efficiency in these cases, it is also possible to include the thin film for controlling transportation of an electron hole or an electron in the lamination.

[0025] Since the JISUCHIRIRU compound illustrated with the above-mentioned structure expression (3) has both electronic transportability ability and electron hole transportability ability, it can be used during a component configuration also as a luminous layer which served as the electron hole transportation layer also as a luminous layer which served both as the electron transport layer. Moreover, it is also possible to consider as the configuration put in the electron transport layer and the electron hole transportation layer by making this JISUCHIRIRU compound into a luminous layer.

[0026] In addition, among drawing 1 and drawing 2 , three are cathode and can use the alloy of a metal [activity / calcium / Li, Mg,] and metals, such as Ag,

aluminum, and In, or the structure which carried out the laminating of these as an electrode material. In the organic electroluminescence devices of a transparency mold, the light transmittance suitable for an application can be obtained by adjusting the thickness of cathode. Moreover, drawing 4 is the closure and a protective layer, and the effectiveness goes up them by making the organic whole electroluminescence devices into wrap structure. A proper ingredient can be used if airtightness is maintained. Moreover, 8 is a drive power source for current impregnation.

[0027] In the organic electroluminescence devices based on this invention, the organic layer has the organic laminated structure (single hetero structure) to which the laminating of an electron hole transportation layer and the electron transport layer was carried out, and said JISUCHIRIRU compound may be used as a formation ingredient of an electron hole transportation layer or an electron transport layer. Or the organic layer has the organic laminated structure (double hetero structure) to which the laminating of an electron hole transportation layer, a luminous layer, and the electron transport layer was carried out one by one, and said JISUCHIRIRU compound may be used as a formation ingredient of a luminous layer.

[0028] When the example of the organic electroluminescence devices which have such an organic laminated structure is shown, drawing 3 has the laminated

structure to which the laminating of organic layer 5a which consists of an anode plate 2, and the electron hole transportation layer 6 and electron transport layer 7 of translucency on the substrate 1 of translucency, and the cathode 3 was carried out one by one, and is the organic electroluminescence devices C of the single hetero structure where a protective coat 4 comes to carry out the closure of this laminated structure.

[0029] In the case of the lamination which omitted the luminous layer as shown in drawing 3 , the luminescence 20 of predetermined wavelength is generated from the interface of the electron hole transportation layer 6 and an electron transport layer 7. These luminescence is observed from a substrate 1 side.

[0030] Moreover, drawing 4 has the laminated structure to which the laminating of organic layer 5b which consists of an anode plate 2, and the electron hole transportation layer 10, the luminous layer 11 and electron transport layer 12 of translucency on the substrate 1 of translucency, and the cathode 3 was carried out one by one, and is the organic electroluminescence devices D of the double hetero structure where a protective coat 4 comes to carry out the closure of this laminated structure.

[0031] In the organic electroluminescence devices shown in drawing 4 , the electron with which the electron hole poured in from the anode plate 2 was poured in from cathode 3 through the electron hole transportation layer 10

reaches a luminous layer 11 through an electron transport layer 12, respectively by impressing direct current voltage between an anode plate 2 and cathode 3. Consequently, the recombination of an electron/electron hole arises in a luminous layer 11, a singlet exciton generates, and luminescence of predetermined wavelength is generated from this singlet exciton.

[0032] In each organic electroluminescence devices C and D mentioned above, the ingredient of light transmission nature, such as glass and plastics, can be suitably used for a substrate 1. Moreover, when using combining other display devices, or when arranging the laminated structure shown in drawing 3 and drawing 4 in the shape of a matrix, it is good considering this substrate as common use. Moreover, Components C and D can all take any structure of a transparency mold and a reflective mold.

[0033] moreover, the anode plate 2 -- a transparent electrode -- it is -- ITO (indium tin oxide) and SnO₂ etc. -- it can be used. Between this anode plate 2 and the electron hole transportation layer 6 (or electron hole transportation layer 10), the thin film which consists of the organic substance or an organometallic compound may be prepared in order to improve the injection efficiency of a charge. In addition, when the protective coat 4 is formed with conductive ingredients, such as a metal, the insulator layer may be prepared in the side face of an anode plate 2.

[0034] Moreover, the electron hole transportation layer 6 and an electron transport layer 7 are organic layers by which the laminating was carried out, the JISUCHIRIRU compound described above to these either or both sides contains organic layer 5a in the organic electroluminescence devices C, and it is good as the luminescent electron hole transportation layer 6 or a luminescent electron transport layer 7. Organic layer 5b in the organic electroluminescence devices D can take various laminated structures, although the electron hole transportation layer 10, the luminous layer 11 containing the above-mentioned JISUCHIRIRU compound, and an electron transport layer 12 are organic layers by which the laminating was carried out. For example, both the electron hole transportation layer, and both [either or] may have a luminescence.

[0035] Moreover, although it is desirable that it is the layer which the electron hole transportation layer 6 or an electron transport layer 7, and a luminous layer 11 turn into from the JISUCHIRIRU compound of this invention especially, these layers may be formed only with said JISUCHIRIRU compound, or you may form by said JISUCHIRIRU compound, other electron holes, or vapor codeposition with electronic transportation ingredients (for example, aromatic amine and pyrazolines etc.). Furthermore, in an electron hole transportation layer, in order to raise electron hole transportability ability, the electron hole transportation layer which carried out the laminating of two or more sorts of electron hole

transportation ingredients may be formed.

[0036] Moreover, in the organic electroluminescence devices C, although a luminous layer may be the electronic transportability luminous layer 7, depending on the electrical potential difference impressed from a power source 8, light may be emitted by the electron hole transportation layer 6 or its interface. Similarly, in the organic electroluminescence devices D, a luminous layer may be an electron transport layer 12 in addition to layer 11, and may be the electron hole transportation layer 10. In order to raise the luminescence engine performance, it is good that it is the structure where the luminous layer 11 which used at least one sort of fluorescence ingredients was made to pinch between an electron hole transportation layer and an electron transport layer. Or the structure where an electron hole transportation layer, an electron transport layer, or both [these] layers were made to contain this fluorescence ingredient may be constituted. In such a case, in order to improve luminous efficiency, it is also possible to include the thin films (a hole blocking layer, exciton generation layer, etc.) for controlling transportation of an electron hole or an electron in the lamination.

[0037] Moreover, you may be the structure in which could use the alloy of a metal [activity / calcium / Li, Mg,] and metals, such as Ag, aluminum, and In, as an ingredient used for cathode 3, and these metal layers carried out the

laminating. In addition, the organic electroluminescence devices corresponding to an application are producible by choosing the thickness and the quality of the material of cathode suitably.

[0038] Moreover, a protective coat 4 acts as closure film, is making the organic whole electroluminescence devices into wrap structure, and can improve charge injection efficiency and luminous efficiency. In addition, if the airtightness is maintained, a single metal or alloys, such as aluminum, gold, and chromium, etc. can choose the ingredient suitably.

[0039] Although the current impressed to each above-mentioned organic electroluminescence devices is usually a direct current, pulse current and an alternating current may be used. If a current value and an electrical-potential-difference value are within the limits which does not carry out component destruction, there will be especially no limit, but when the power consumption and the life of organic electroluminescence devices are taken into consideration, it is desirable to make light emit efficiently with as small electrical energy as possible.

[0040] Next, drawing 5 is the example of a configuration of the flat-surface display which used the organic electroluminescence devices of this invention. In the full color display, like illustration, red (R) and the green organic layer 5 (5a, 5b) which can emit light in the three primary colors of (G) and blue (B) are

allotted between cathode 3 and an anode plate 2. It can prepare in the shape of [which crosses mutually] a stripe, it is chosen by the luminance-signal circuit 14 and the control circuit 15 with a built-in shift register, and a signal level is impressed to each, and cathode 3 and an anode plate 2 are constituted so that the organic layer of the location (pixel) where the cathode 3 and the anode plate 2 which were chosen by this cross may emit light.

[0041] That is, it is a 8x3RGB simple matrix, and drawing 5 arranges the layered product 5 which consists of one side between cathode 3 and an anode plate 2, even if there are few electron hole transportation layers, and luminous layers and electron transport layers either (refer to drawing 3 or drawing 4). Both cathode and an anode plate are made to intersect perpendicularly in the shape of a matrix mutually, impress a signal level serially by the control circuits 15 and 14 with a built-in shift register, and they are constituted so that light may be emitted in the decussation location, while carrying out patterning to the shape of a stripe. Of course, the EL element of this configuration can be used also as picture reproducer as a display of an alphabetic character, a notation, etc. Moreover, the stripe-like pattern of cathode 3 and an anode plate 2 is arranged for every color of red (R), green (G), and blue (B), and it becomes possible to constitute multicolor or all full color solid-state mold flat-panel displays.

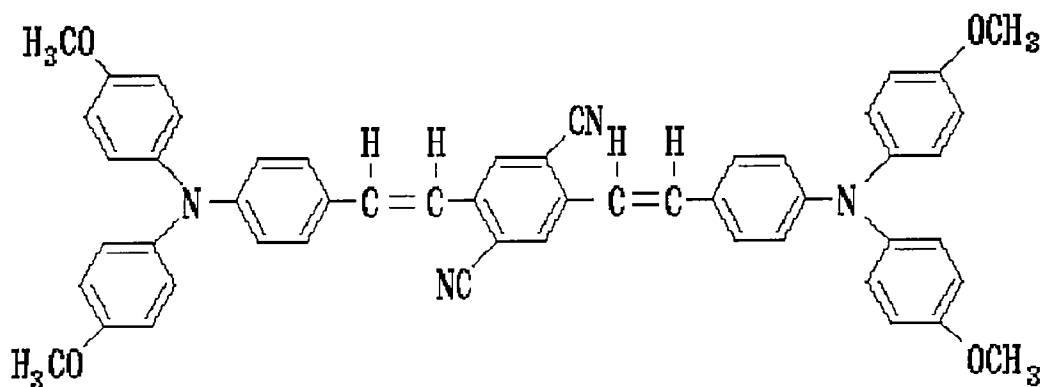
[0042]

[Example] Hereafter, although this invention is concretely explained about an example, this invention is not limited to the following examples.

[0043] Example 1 this example is R1 among the above-mentioned JISUCHIRIRU compounds of a general formula (1), R2, and R3 and R4. It is 3-methoxypheny radical R6 and R8 It is the example which produced the organic electroluminescence devices of single hetero structure, using the compound with a cyano group of following structure-expression (3)-1 as an electron hole transportability luminescent material.

[Formula 4]

構造式 (3) - 1 :



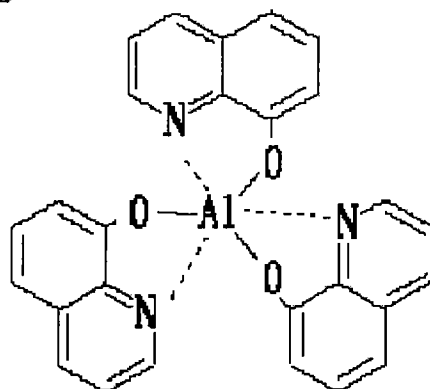
[0044] First, the 30mmx30mm glass substrate with which the anode plate which consists of ITO with a thickness of 100nm was formed on one front face into the vacuum evaporation system was set. The substrate was approached, the metal mask which has two or more 2.0mmx2.0mm unit openings as a vacuum evaporation mask has been arranged, and the compound of above-mentioned

structure-expression (3)-1 was formed as an electron hole transportation layer (*****) in thickness of 50nm under the vacuum of 10^{-4} or less Pa with the vacuum deposition method. The vacuum evaporation rate was carried out in 0.1nm/second.

[0045] Furthermore, Alq3 (tris (eight quinolinol) aluminum) of the following structure expression was vapor-deposited in contact with the electron hole transportation layer as an electronic transportation ingredient. Alq3 from -- thickness of this becoming electron transport layer was also set to 50nm, and the vacuum evaporation rate was carried out in 0.2nm/second.

[Formula 5]

Alq₃ :



[0046] The cascade screen of Mg and Ag was adopted as a cathode material, and by vacuum evaporation, this was also formed in the thickness of 50nm (Mg film) and 150nm (Ag film) as vacuum evaporation rate 1nm/a second, and produced the **** organic electroluminescence devices shown in drawing 3 by

the example 1.

[0047] Thus, forward bias direct current voltage was applied to the organic electroluminescence devices of the produced example 1 under nitrogen-gas-atmosphere mind, and the luminescence property was evaluated. The luminescent color was red, and as a result of performing spectrometry, as shown in drawing 6 , it obtained the spectrum which has a luminescence peak in 650nm. The spectroscopy which used the photodiode array by the Otsuka electronic company as the detector was used for spectrometry. Moreover, when the electrical-potential-difference-measurement of luminance is performed, as it is shown in drawing 8 , they are 1200 cd/m² at 9.5V. Brightness was obtained.

[0048] After producing these organic electroluminescence devices, although it was left for one month under nitrogen-gas-atmosphere mind, component degradation was not observed. Moreover, initial brightness 200 cd/m² The current value was energized uniformly and continuation luminescence was carried out, and when carrying out forcible degradation, it was 1000 hours until brightness was halved.

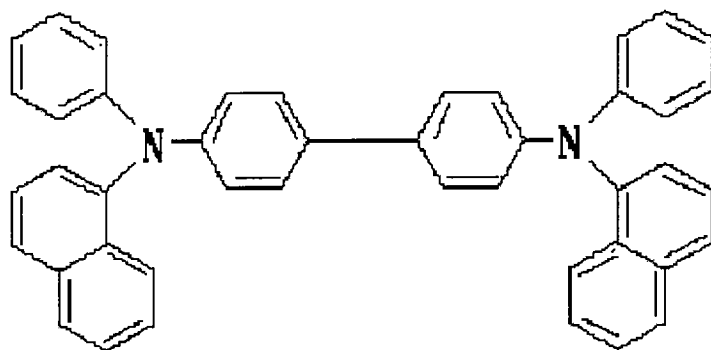
[0049] Example 2 this example is R1 among the above-mentioned JISUCHIRIRU compounds of a general formula (1), R2, and R3 and R4. It is 3-methoxyphenyl radical R6 and R8 It is the example which produced the organic electroluminescence devices of single hetero structure, using the compound

with a cyano group of above-mentioned structure-expression (3)-1 as an electronic transportability luminescent material.

[0050] First, the 30mmx30mm glass substrate with which the anode plate which consists of ITO with a thickness of 100nm was formed on one front face into the vacuum evaporation system was set. The substrate was approached, the metal mask which has two or more 2.0mmx2.0mm unit openings as a vacuum evaporation mask has been arranged, and alpha-NPD (alpha-naphthylphenyl diamine) of the following structure expression was formed as an electron hole transportation layer in thickness of 50nm under the vacuum of 10^{-4} or less Pa with vacuum evaporation technique. The vacuum evaporation rate was carried out in 0.1nm/second.

[Formula 6]

α -NPD :



[0051] Furthermore, the compound of above-mentioned structure-expression

(3)-1 was vapor-deposited in contact with the electron hole transportation layer as an electronic transportation ingredient. The above-mentioned structure expression (3) Thickness of the electron transport layer (*****) which consists of a compound of -1 was also set to 50nm, and the vacuum evaporation rate was carried out in 0.2nm/second.

[0052] The cascade screen of Mg and Ag was adopted as a cathode material, and by vacuum evaporation, this was also formed in the thickness of 50nm (Mg film) and 150nm (Ag film) as vacuum evaporation rate 1nm/a second, and produced the **** organic electroluminescence devices shown in drawing 3 by the example 2.

[0053] Thus, forward bias direct current voltage was applied to the organic electroluminescence devices of the produced example 2 under nitrogen-gas-atmosphere mind, and the luminescence property was evaluated. The luminescent color was red, and as a result of performing spectrometry like an example 1, as shown in drawing 7 , it obtained the spectrum which has a luminescence peak in 650nm. Moreover, when the electrical-potential-difference-measurement of luminance is performed, as it is shown in drawing 9 , they are 600 cd/m² at 10.5V. Brightness was obtained.

[0054] After producing these organic electroluminescence devices, although it was left for one month under nitrogen-gas-atmosphere mind, component

degradation was not observed. Moreover, initial brightness 200 cd/m² The current value was energized uniformly and continuation luminescence was carried out, and when carrying out forcible degradation, it was 700 hours until brightness was halved.

[0055] Example 3 this example is R1 among the above-mentioned JISUCHIRIRU compounds of a general formula (1), R2, and R3R4. It is 3-methoxyphenyl radical R6 and R8 It is the example which produced the organic electroluminescence devices of double hetero layer structure, using the compound with a cyano group of above-mentioned structure-expression (3)-1 as a luminescent material.

[0056] First, the 30mmx30mm glass substrate with which the anode plate which consists of ITO with a thickness of 100nm was formed on one front face into the vacuum evaporation system was set. The substrate was approached, the metal mask which has two or more 2.0mmx2.0mm unit openings as a vacuum evaporation mask has been arranged, and alpha-NPD of the above-mentioned structure expression was formed as an electron hole transportation layer in thickness of 30nm under the vacuum of 10⁻⁴ or less Pa with the vacuum deposition method. The vacuum evaporation rate was carried out in 0.2nm/second.

[0057] Furthermore, the compound of above-mentioned structure-expression

(3)-1 was vapor-deposited in contact with the electron hole transportation layer as a luminescent material. The above-mentioned structure expression (3) Thickness of the luminous layer which consists of a compound of -1 was also set to 30nm, and the vacuum evaporation rate was carried out in 0.2nm/second.

[0058] Furthermore, it is Alq3 of the above-mentioned structure expression as an electronic transportation ingredient. It vapor-deposited in contact with the luminous layer. Alq3 Thickness was set to 30nm and the vacuum evaporation rate was carried out in 0.2nm/second.

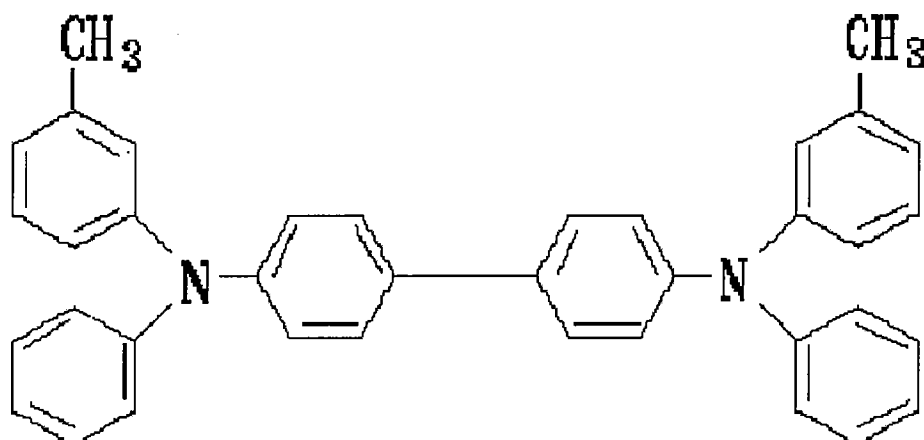
[0059] The cascade screen of Mg and Ag was adopted as a cathode material, and by vacuum evaporation, this was also formed in the thickness of 50nm (Mg film) and 150nm (Ag film) as vacuum evaporation rate 1nm/a second, and produced the **** organic electroluminescence devices shown in drawing 4 by the example 3.

[0060] Thus, forward bias direct current voltage was applied to the organic electroluminescence devices of the produced example 3 under nitrogen-gas-atmosphere mind, and the luminescence property was evaluated. The luminescent color was red, and as a result of performing spectrometry, it obtained the spectrum which has a luminescence peak in 650nm. When the electrical-potential-difference-measurement of luminance was performed, they are 1800 cd/m² at 8.5V. Brightness was obtained.

[0061] After producing these organic electroluminescence devices, although it was left for one month under nitrogen-gas-atmosphere condition, component degradation was not observed. Moreover, initial brightness 200 cd/m² The current value was energized uniformly and continuation luminescence was carried out, and when carrying out forcible degradation, it was 1500 hours until brightness was halved.

[0062] Changed to alpha-NPD as an example 4 electron-hole transportability ingredient, and TPD (triphenyl diamine derivative) of the following structure expression was used, and also lamination and the forming-membranes method produced organic electroluminescence devices based on the example 2.

TPD :



[0063] The organic electroluminescence devices of this example also presented luminescence of the same red as an example 2. The spectrum was in

agreement with the spectrum of the organic electroluminescence devices of an example 2 as a result of spectrometry.

[0064]

[Function and Effect of the Invention] Since the JISUCHIRIRU compound expressed with said organic layer by said general formula (1) is contained in the organic electroluminescence devices by which the organic layer which has a luminescence field is prepared between an anode plate and cathode according to the organic electroluminescence devices of this invention, it becomes possible to offer the organic electroluminescence devices which have stable red luminescence by high brightness.

DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

[Drawing 1] It is the important section outline sectional view of the organic electroluminescence devices based on this invention.

[Drawing 2] They are other important section outline sectional views of **** organic electroluminescence devices.

[Drawing 3] They are other important section outline sectional views of ****

organic electroluminescence devices.

[Drawing 4] They are other important section outline sectional views of ****

organic electroluminescence devices.

[Drawing 5] It is the block diagram of the full color flat-surface display using ****

organic electroluminescence devices.

[Drawing 6] It is the emission spectrum Fig. of the organic electroluminescence devices by the example 1 of this invention.

[Drawing 7] It is the emission spectrum Fig. of the organic electroluminescence devices by the **** example 2.

[Drawing 8] It is the electrical-potential-difference-brightness property Fig. of the organic electroluminescence devices by the **** example 1.

[Drawing 9] It is the electrical-potential-difference-brightness property Fig. of the organic electroluminescence devices by the **** example 2.

[Description of Notations]

1 [-- A protective coat, 5, 5a, 5b / -- An organic layer, 6 / -- An electron hole transportation layer, 7 / -- An electron transport layer, 8 / -- A power source, 10 / -- An electron hole transportation layer, 11 / -- A luminous layer, 12 / -- An electron transport layer, 14 / -- A luminance-signal circuit, 15 / -- A control circuit, 20 / -- Luminescence light, A B C, D / -- Organic electroluminescence devices] -- A substrate, 2 -- A transparent electrode (anode plate), 3 -- Cathode, 4

Theismann, Paula M.

From: Moshe, Mirella
Sent: Wednesday, April 25, 2007 1:53 PM
To: Theismann, Paula M.
Subject: FW: Risperidone - WO 2005/030772

can you please check and let me know? (status, national filing, US counterparts)

thanks.

matter no. is 20065860-0053

From: Adva.Shickartzi@teva.co.il [mailto:Adva.Shickartzi@teva.co.il]
Sent: Wednesday, April 25, 2007 2:50 AM
To: Moshe, Mirella; Saito, Marina N.
Subject: Fw: Risperidone - WO 2005/030772

Dear Mirella and Marina,

Kindly update me as to the status of the following publications;

WO 05/030772
WO 04/092143
WO04/ 0209898

Regards,
Adva Shickartzi

Adva Shickartzi
Patent Department
Teva Pharmaceutical Industries Ltd.

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----- Forwarded by Adva Shickartzi/PTV/TEVA/IL on 25/04/2007 09:49 -----

From: "Saito, Marina N." <msaito@sonnenschein.com>
Sent To: <Adva.Kubani@teva.co.il>
CC: "Moshe, Mirella" <mmoshe@sonnenschein.com>
BCC:
Subject: Risperidone - WO 2005/030772
Delivery date: 03/01/2007

4/25/2007

Dear Adva,

No U.S. applications have yet been filed for WO 2005/030772.

We will provide you with another update next month. In the meantime, please let us know if you have any questions regarding this matter.

Best Regards,

Marina

Marina N. Saito
Sonnenschein Nath & Rosenthal LLP
7800 Sears Tower
Chicago, IL 60606
Direct: 312-876-8157
Fax: 312-876-7934

Link to document: [Link](#)

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4/25/2007

Theismann, Paula M.

From: Moshe, Mirella
Sent: Wednesday, April 25, 2007 3:47 PM
To: Theismann, Paula M.
Subject: RE: Risperidone - WO 2005/030772

Paula,
Thanks.

For the '772 application - when you checked it as part of the monitoring table, you noted also New Zealand and South Africa national filings, and did not note Canada. Can you please clarify?

Also - can you please advise the national stage deadline for '143 and confirm that there are no US corresponding applications for either '772 or '143 (not only through national stage).

From: Theismann, Paula M.
Sent: Wednesday, April 25, 2007 4:18 PM
To: Moshe, Mirella
Subject: RE: Risperidone - WO 2005/030772

Hi Mirella

Attached are Dialog Derwent/Inpadoc search results for WO 200530772 and WO 2004092143.

I could not locate anything for WO 20040209898 - may be an incorrect number.

Re WO 200530772 - National stage entered in EP 2004787585 published as EP 1670797.
Australia Ser. No. 2004276092
Canada Ser. No. 2540360

Priority Application - Indian Ser. No. 2003DEI209 - published as 200301209

Re WO 2004 092143 - National stage does not appear to have been entered in India only - India 200300914. Australia preassigns serial numbers and note publication date is within a few days of the WO publication date - could not locate anything to confirm entrance to national stage in Australia. Priority application is the WO application - WO 2003IN157.

No national stage filings in US for either application.

Thank you.

Paula

4/25/2007

Paula M. Theismann
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Subject: FW: Risperidone - WO 2005/030772

can you please check and let me know? (status, national filing, US counterparts)

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Dear Mirella and Marina,

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WO04/ 0209898

Regards,
Adva Shickartzi

Adva Shickartzi
Patent Department
Teva Pharmaceutical Industries Ltd.

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Marina

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